An Advanced Open Path

Atmospheric Pollution Monitor for Large Areas*

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Introduction

Over 100 million gallons of radioactive and toxic waste materials generated in weapon materials production are stored in 322 tanks buried within large areas at DOE sites. Toxic vapors occur in the tank headspaces due to the solvents used and chemical reactions within the tanks. To prevent flammable or explosive concentrations of volatile vapors, the headspace are vented, either manually or automatically, to the atmosphere when the headspace pressure exceeds preset

* Research sponsored by the US Department of Energy's Morgantown Energy Technology Center, under contract DE-AR21-95MC32087 with Northrop Grumman, 1350 Beulah Road, Pittsburgh, PA 15235; fax: (412) 256-1661.

values. Furthermore, 67 of the 177 tanks at the DOE Hanford Site are suspected or are known to be leaking into the ground.[1]

Objectives

These underground storage tanks are grouped into "tank farms" which contain closely spaced tanks in areas as large as 1 km^2. *The objective of this program is to protect DOE personnel and the public by monitoring the air above these tank farms for toxic air pollutants without the Monitor entering the tanks farms which can be radioactive.* A secondary objective is to protect personnel by monitoring the air above buried 50 gallon drums containing moderately low radioactive materials but which could also emit toxic air pollutants.

Approach

Our approach is to combine two technologies to take three measurements: a pulsed CO_2 laser is used to measure (1) absorption spectra in the 9-11 μm spectral region and (2) the distance over which the measurements are made. An acousto-optic tunable filter (AOTF) is used to measure (3) thermal emission spectra in the 3-14 μm spectral region. This combination is the thermal emission, laser absorption (TELA) air pollution monitor concept described herein.[2]

More explicitly, for long open-path remote sensing and quantitative measurements of atmospheric concentrations of trace vapors, differential-absorption lidar (DIAL) is the most sensitive technique. In this technique, the laser is tuned to the absorption peak of a pollutant vapor and then to a nearby wavelength at which the pollutant does not absorb. *Infrared* DIAL systems are preferred because they are sensitive to the laser energy, are relatively "eye safe", and can operate in relatively poor weather. Furthermore, *CO_2 DIAL systems* are preferred because they have sufficient power for measurements over multi-kilometer distances and because their 9-11 μm spectral coverage is in the infrared "fingerprint region" of 8-14 μm where most molecule-specific absorption lines occur.[3]

DIAL systems can also measure the distance over which the measurements are made -- *without retroreflectors*. However, because not all of the molecules of interest absorb in the 9-11 μm spectral region, a CO_2 DIAL system must be complemented with a system which covers a broader range of wavelengths. The AOTF is a good choice for the complementary system because it:

1. Is easily integrated into a DIAL system
2. Monitors thermal emission spectra passively
3. Covers a broad wavelength region of 3-14 μm
4. Can be quickly tuned to any desired wavelength
5. Is very sensitive to narrow lines by measuring their derivatives
6. Has high reliability since it does not have any moving mechanical parts
7. Provides complete images which can be used with focal plane array detectors.

To maintain high signal-to-noise ratios (SNR) the last feature is not used in the present system; single point detectors are used. However, the imaging capability could be added at a later time by replacing the point detectors with detector arrays and modifying the detector imaging optics.

Project

System Description

The TELA air pollution monitor development program is at the end of the design phase. The basic system is shown schematically in Figure 1. It is comprised of seven key elements: a CO_2 laser, a laser beam expander, telescope, matching optics, an AOTF, detectors, and computers. The commercial CO_2 laser, composed of an oscillator and an amplifier, is electronically tunable over ~90 lines in the 9.2-10.9 μm spectral region. It operates at 5 kHz 1.5 mJ/pulse on the highest gain line. This energy is sufficient to monitor ranges up to 4 km. The pulse width of the linearly polarized beam is 100 ns or longer, depending on the laser line. The output power of the laser is measured by reflecting from a ZnSe window a small portion of the beam to a room temperature HgCdTe detector optimized for 10.6 μm. This detector is shown in the optical bench layout in Figure 2 as the small object near the bottom of the laser oscillator. The larger rectangle below the detector is a HeNe alignment laser.

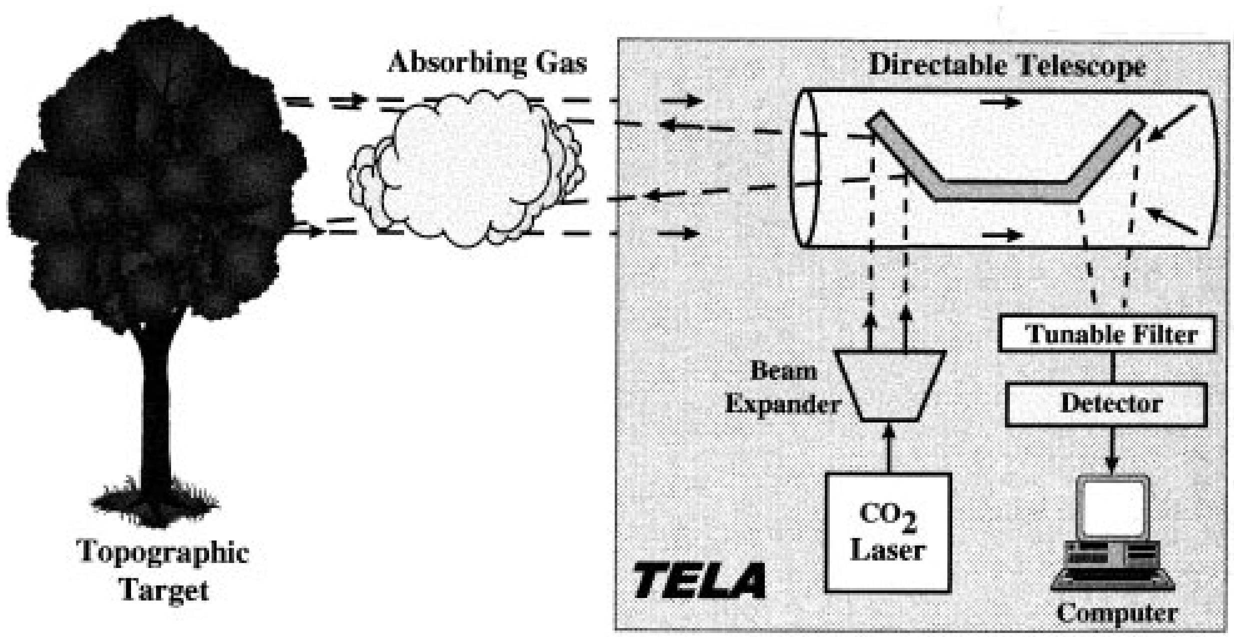

Figure 1. The TELA monitor augments an active CO_2 lidar system with a passive AOTF system.

A beam expander increases the laser beam diameter from 0.44 cm to 6.6 cm. The larger beam is needed for eye safety. Also, it samples a larger volume of the atmosphere. A TV camera monitors the outdoor scene by using the reflection of the scene from the Ge lens surface at the output of the beam expander. A Cassegrainian telescope with a 12.7 cm output mirror directs the laser beam to a steering mirror on a pan & tilt stage which provides a 75° horizontal and 20° vertical field of regard with a 0.004° resolution. The radiation collected by the 31.5 cm input mirror of the Cassegrainian telescope is thermal emission from the atmosphere which may or may not include a reflected laser beam. This beam has its diameter reduced and is collimated before it enters the AOTF.

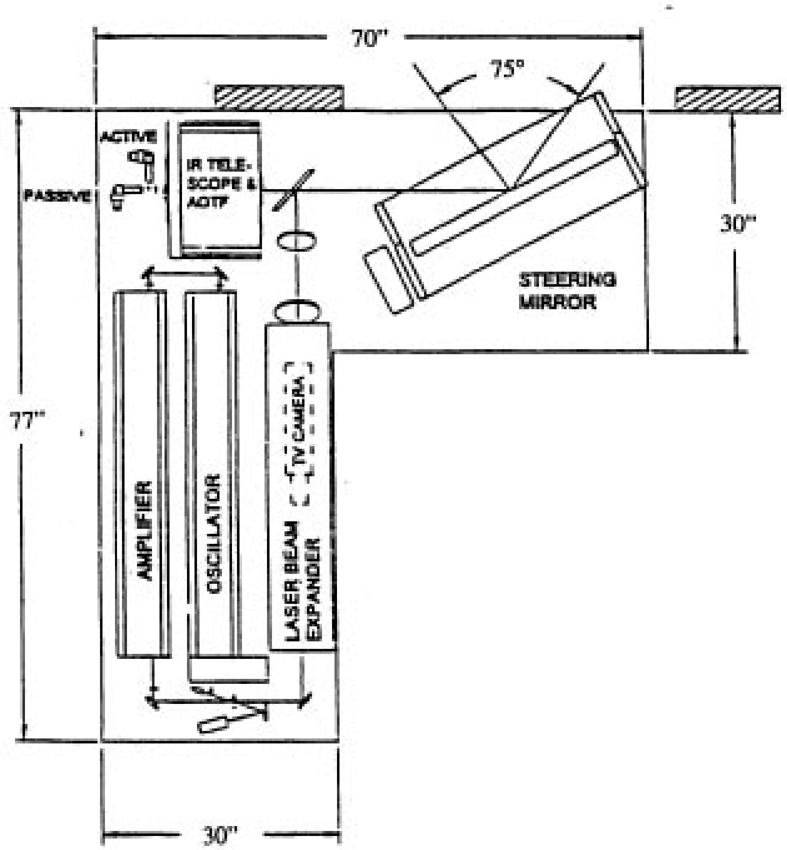

Figure 2. The optical bench layout (dimensions are in inches) is compact with all ancillary equipment under the bench.

Operation of the AOTF is shown schematically in Figure 3. The transducer, electrically excited with less than 5 W of power, transforms the electromagnetic rf waves into acoustic waves which propagate into the AO crystal. These bulk acoustic waves force the material's indices of refraction into a spatial pattern resembling a diffraction grating. This grating structure diffracts a narrow spectral band of the incident radiation into the first order (since the AO crystal is a birefringent material, diffraction into higher orders is not allowed). The acceptance angle, nominally between 15°

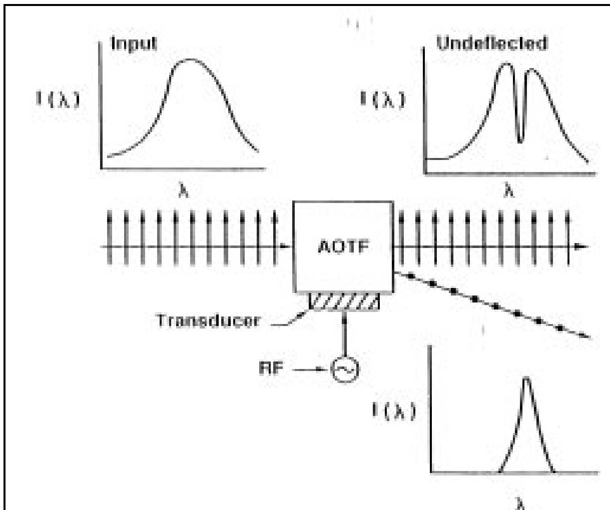

Figure 3. AOTFs are electronic solid state filters.

and 60°, and the spectral width of the diffracted beam are determined by the AOTF design. The diffracted beam has its plane of polarization rotated 90° and has its propagation direction changed a few degrees. Thus, the AOTF is a continuously adjustable narrow-band optical filter which operates on one polarization only.

Two beams emerge from the AOTF: the filtered, or diffracted beam, and the unfiltered beam. Two sets of matching optics direct the beams to 0.75 mm square HgCdTe detectors which are cooled by Stirling coolers to 77 K. The unfiltered beam, containing the laser pulse if present, is passed through a 9-11 μm cold filter in front of the "active" detector. The diffracted beam has a fixed passband of $3\ cm^{-1}$ with its center wavelength, uniquely determined by the frequency of the acoustic beam, located anywhere between 3 and 14 μm. It is directed to the "passive" detector.

Two computers are used. The control computer operates the monitor and processes signals from the three detectors. The analysis computer sets up the data collection process. The operator enters the gases to be measured, line-of-sight angular specifications for collection, times of collection. The operator has continuous visual coverage of the line-of-sight scene through a 8" TV monitor. The analysis computer transmits the operating parameters to the control computer. It also analyzes, in real time, the information received from the control computer. The results are output to a 17" color monitor and put into archival storage.

To monitor several tank farms, the TELA Monitor is mobile and self-contained. The entire Monitor fits into a light truck or a small recreational vehicle (RV) such as the one illustrated in Figure 4. This mobility requires additional instrumentation. The analysis computer records the location and altitude of the Monitor using a Global Positioning System (GPS) receiver. The compass heading of the telescope is recorded as given by a meteorological station which also provides the air temperature, relative humidity, and wind velocity.

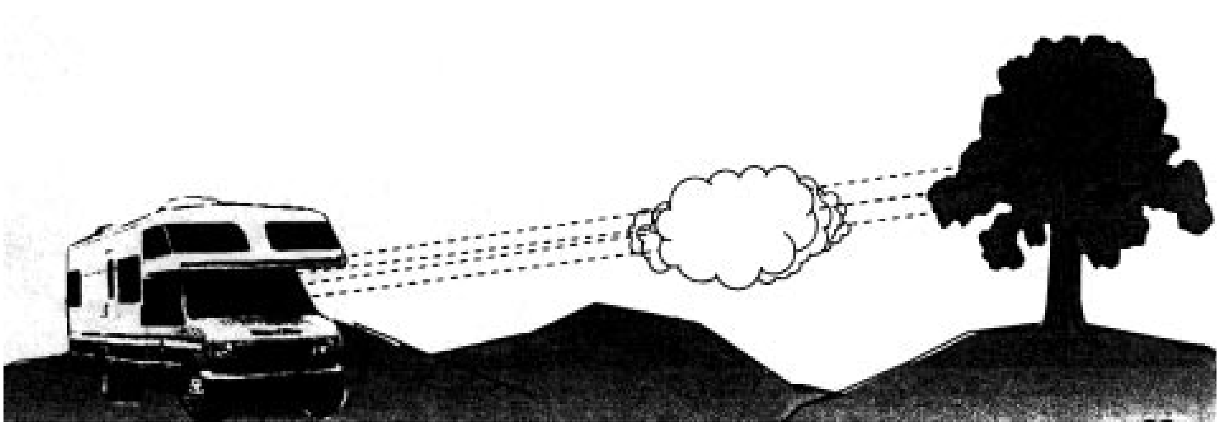

Figure 4. The TELA Monitor can be placed in a small RV and taken anywhere.

System Operation

Before measurements are taken, the operator must set several parameters by responding to user-friendly software. The analysis computer uses Microsoft Windows to query for the parameters, starting with the laser repetition rate and plume thickness. The plume thickness is useful if there is reasonable expectation that any high concentration of a trace gas is coming from a known source such as a vent pipe or smoke stack. The trace gases to be measured are then selected from a table presented on the computer screen. Up to twenty angular settings of the steering mirror are entered. Finally, the data collection parameters are entered: start time, stop time, and time between data sets. A data set is one complete cycle of the specified telescope settings with all specified trace gases being measured at each telescope setting. Manual operation of the system is also permitted.

The operator-specified parameters are passed to the control computer which governs the electrical and mechanical operation of the system. The first action is to set, via stepping motors, the steering mirror to the first specified setting. A 5 kHz maximum timing circuit is then adjusted to provide trigger pulses, at the specified laser repetition rate, to a Pockels cell in the laser oscillator. Based on the measurement algorithm for each gas, the control computer receives (from the analysis computer) a table of wavelengths for both the laser absorption and thermal emission measurements. The laser oscillator wavelength is controlled by an accurately rotated grating on a galvanometer. The AOTF wavelength is controlled by a frequency synthesizer. After performing all the gas concentration measurements, the telescope setting is changed and the entire process repeated.

The electrical signals from all three detectors are amplified and sent to the control computer. The output data sequence is illustrated in Figure 5. The transmitted laser pulse and the received laser pulse are both digitized at a 100 Mhz rate and processed by a digital signal processor (DSP). The transmission and received times of the pulses are determined from their leading edges. The time of flight of the pulse to any reflecting object, e.g., the tree shown in Figure 1, is measured by the time between the leading edges of the pulses. This interval determines the distance, or range, to that object (to within ±1.5 m) up to 4 km. The range is needed to determine the average concentration of a vapor since the spectral measurements actually give the product of concentration and range.

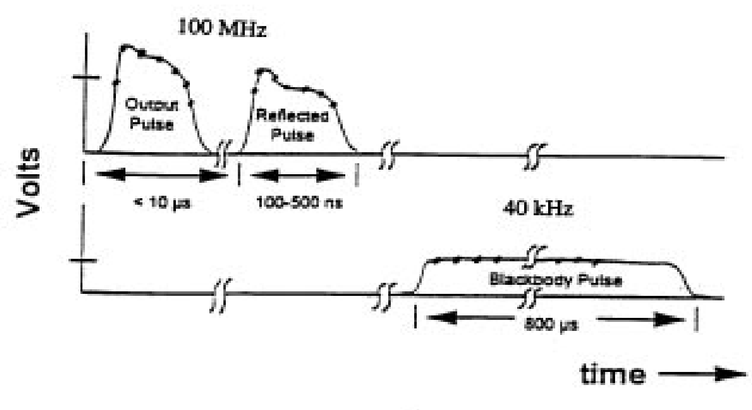

Figure 5. Digitization of the detector pulses provides versatility and accuracy.

The thermal emission pulse is measured between laser pulses to prevent scattered laser radiation from giving false readings. Since the thermal emission pulse is very much longer than the laser pulses, it is digitized at a slower rate: 40 kHz.

The control computer calculates the power in each pulse. The ratio of laser power transmitted (from the room temperature detector) to laser power received (from the active detector) is computed and passed to the analysis computer. The thermal emission power (from the passive detector) is also transmitted to the analysis computer. These processes are repeated for the number of pulses specified by the analysis computer via its measurement algorithms. The analysis computer averages the received measurements for each gas, thereby increasing the signal-to-noise ratio and reducing the variance.

Three gas sets are measured for each telescope setting. The first set is composed of six normal heteronuclear atmospheric constituents: H_2O, CO_2, CH_4, N_2O, O_3, and CO. The second set contains non-toxic air pollutants and the third set toxic air pollutants. For each set the measured concentrations are output to the 17” color monitor in the form of a bar chart, as shown in Figure 6 for a representative third set. The station number at the top of the chart is the telescope setting number (previously specified by the operator) and the range is the measured distance from the TELA Monitor to the reflecting target. Each gas is identified on the abscissa and the light gray bar* gives the concentration assuming that it is evenly distributed over the measurement path length. The dark gray bar gives the concentration assuming that all pollutants are located in the plume thickness specified by the operator. The white bar is the danger level. It is the time-weighted average (TWA) threshold limit value in ppm by volume at which nearly all workers may be exposed, day after day, for a normal 8-hour workday and 40-hour workweek, without any adverse effects.[4]

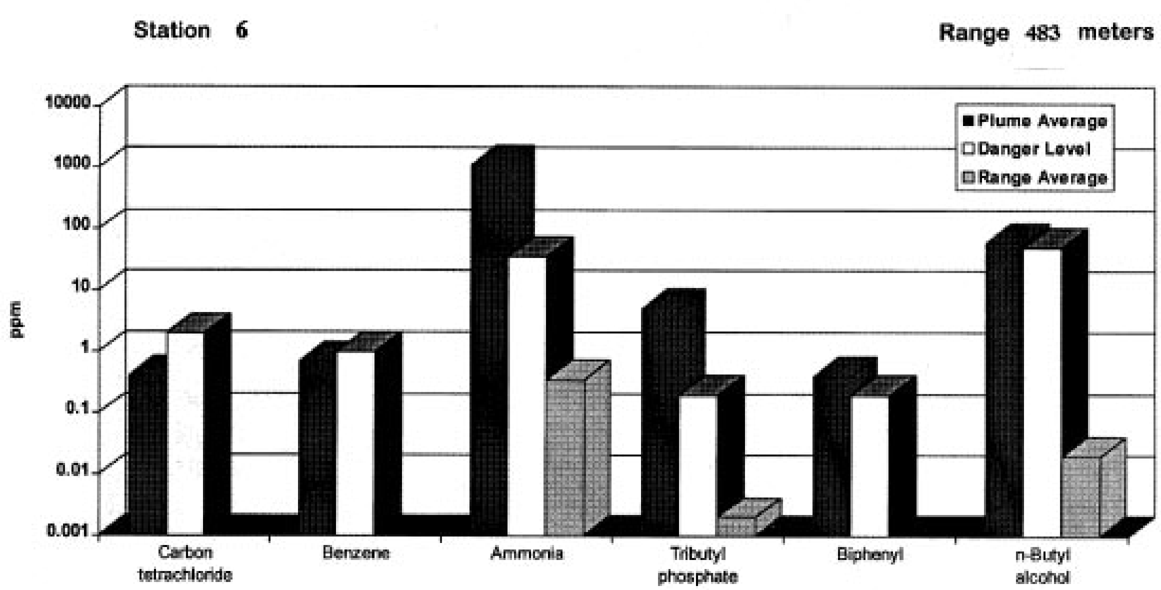

Figure 6. Measured results are displayed in an easily understood graphical form.

* On a color monitor the light gray is a bright blue, the dark gray is a bright green, and the white is a bright red.

A special operating feature of an AOTF is its ability to measure derivatives of spectral lines which are narrower than its passband. This feature can be seen from Figure 7. When the acoustic frequency is modulated, the passband oscillates around a narrow spectral line as shown, yielding an amplitude modulated (AM) output signal. If the line is broader than the AOTF passband, there will be little AM signal. If the background radiation is broad, e.g., blackbody radiation, the signal-to-background ratio will be dramatically increased (as high as three or four orders of magnitude[5]) although the SNR will be decreased. This technique is well known in tunable diode lasers[6] and is most useful for detecting light molecules because of their narrow lines.

The TELA Monitor determines the spectra derivative numerically. With only the AOTF in operation, the passive detection system can monitor an emission line for an arbitrarily long time. At the 40 kHz digital sampling rate, the center wavelength of the AOTF is stepped through a series of wavelengths, with the sequence being repeated as long as the emission line is being measured. The average of the measured value at each wavelength is then determined and the derivative calculated numerically.

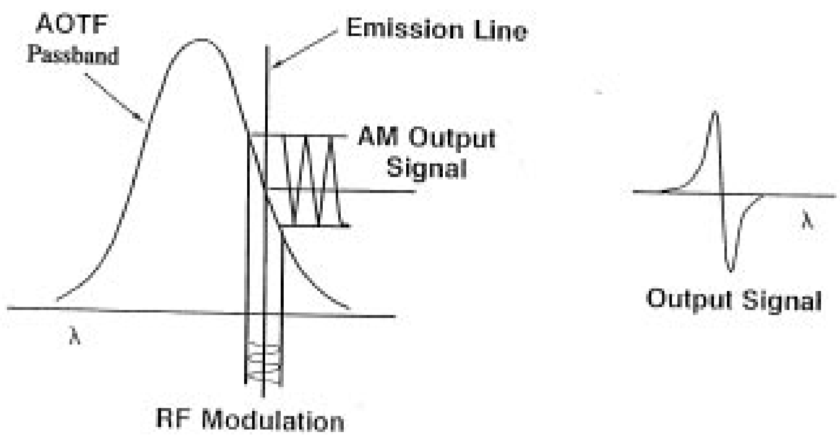

Figure 7. Spectral derivatives are easily obtained with an AOTF.

System Features

The combination of the active and passive detection systems provides features which either system alone does not have. The way these two detection system complement each other is shown in Table 1. The AOTF has the broadest wavelength coverage with the shortest wavelength determined by the fact that HF is the only heteronuclear molecule which has any strong lines shorter than this wavelength.[7] The longest wavelength is determined by the maximum sensitivity of commercial HgCdTe detectors

Table 1. Active and passive systems complement each other.

Feature	Active	Passive
Measures Range	**Yes**	no
Shortest Wavelength	9 μm	**3 μm**
Longest Wavelength	11 μm	**14 μm**
Measures Derivatives	no	**Yes**
Imaging Capability	no	**Yes**
High Line Selectivity	**Yes**	no
Poor Weather Operation	**Yes**	no

and by the atmospheric transmission window which cuts off rather abruptly at 13.5 μm. Furthermore, the AOTF can easily determine spectral derivatives and produces whole images. The CO_2 lidar system measures distances to reflecting objects, has very high line selectivity due to its very narrow linewidth, and can operate in relatively poor weather due to its high intensity and long wavelengths.

Accomplishments

The AOTF has been designed (see Figure 8) and fabricated (see Figure 9) from Tl_3AsSe_3, a very good material for the far infrared. The input aperture is 1.5 cm by 1.0 cm with a 1.0 cm square output aperture. The crystal is 5.4 cm long with a 4.2 cm long lithium niobate transducer. For an efficient and uniform interaction between the acoustic and optic beams, the AOTF is designed so that the acoustic beam is not appreciably absorbed by the crystal. The lead absorbs the acoustic beam which is not absorbed by the crystal. The lead absorption prevents excessive heating of the crystal and stops the acoustic beam from bouncing around inside the crystal where it could again interact, in a deleterious way, with the optical beam. The first and zero order output beams are separated by 10.6° which makes their angular separation very easy.

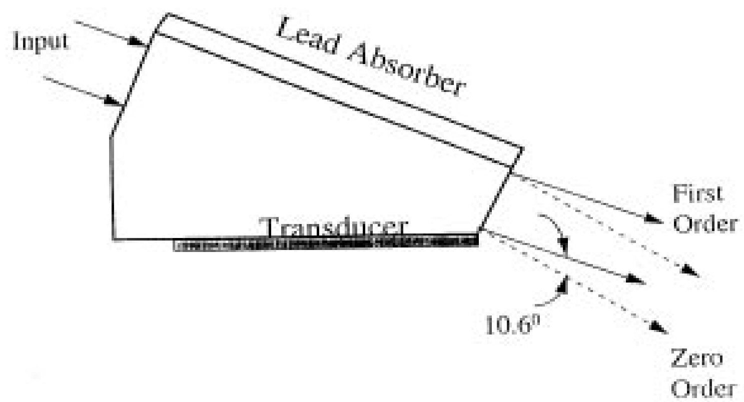

Figure 8. The AOTF design produces filtered (first order) and unfiltered (zero order) beams which are easily separated by their angular difference.

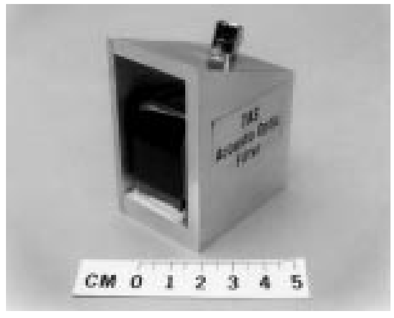

Figure 9. The fabricated AOTF is shown in its aluminum holder. The top protrusion is the electrical connection to the transducer.

The Los Alamos National Laboratory has developed a relevant simulation code: CO_2 Simulation and Optimization Numerics for DIAL (SONDIAL). It has been applied to the TELA Monitor overseeing a simulated tank farm with a 1.8 m square 80% reflecting copper foil covered canvas draped over the far fence as the reflecting target. The range was set at 1.6 km, thus allowing the Monitor (with its 75° field of regard) to sense the air above the entire surface of the simulated 1 km x 1 km tank farm. With typical system parameters, and sampling the absorption at each wavelength 100 or more times to increase the SNR by a factor of ten, the TELA Monitor will be able to measure an absorption of 0.3%. The SNR is speckle limited to range of 2.1 km but is sufficiently high that measurements can be taken out to 4 km.

We used the SONDIAL code to simulate concentration measurements by the TELA Monitor of the toxic air pollutants listed in Table 2. These toxic pollutants have some of the highest concentrations measured in the Hanford tank headspaces. The simulations assume that the pollutant plume is as wide and as high as the 1.8 m square reflector. The detectable plume thickness is the smallest thickness which could be detected if the chemical density in the plume was the same as that in the average tank headspace (the actual plume thickness will depend on the wind and atmospheric conditions). Ammonia and Tributyl phosphate will be easily detected; n-Butyl alcohol, Trichloroflouromethane, and Ethanol will be detected in slightly thicker plumes. Ammonia is present in all the tanks and Tributyl phosphate in 44% of the tanks. These vapors are therefore good tag chemicals to indicate that a tank is leaking. They are also the only two chemicals with headspace concentrations which exceed the TWA exposure limits.

Table 2. Toxic air pollutants found at high concentrations in the Hanford tank headspaces which are detectable by the CO_2 lidar system.

Chemical	TWA (ppm)	Average Headspace Density (ppm)	Detectable Plum Thickness (m)
Ammonia	35	223	0.0003
n-Butyl Alcohol	50	2.80	.24
Butyraldehyde	40	1.33	2.7
Acetone	750	0.86	38
Acetonitrile	40	0.57	8.8
Ethanol	1000	0.43	7.5
Tributyl phosphate	0.2	0.30	0.09
Trichloroflouromethane	800	0.19	0.41
Hexane	50	0.13	81
Acetaldehyde	100	0.09	38
1,3-Butadiene	1000	0.05	18
Benzene	1	0.04	38
Ethylene oxide	1	0.03	312

Benefits

A major benefit relative to other monitoring technologies is the large area which is monitored from a distance. This advantage is illustrated in Figure 10 in which the circles denote nominal operating ranges of point samplers, Fourier Transform Infrared (FTIR) Spectrometers, and the TELA Monitor. Superimposed on these circles of coverage is the Hanford 200-East tank farms, drawn to the same scale as the circles. As shown, the TELA Monitor is positioned at a corner of the farm and views the entire area — without having to enter the tank farm properties, thus avoiding the necessity of having the operator wear special protective coating and/or risking contamination of the TELA Monitor.

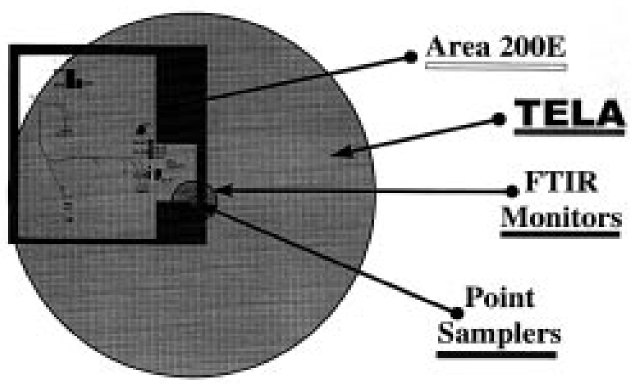

Figure 10. The TELA Monitor can survey several tank farms from a single location without entering them (stripes shown in circle are a reproduction artifact).

There are many other benefits associated with the TELA Monitor. A list of the major benefits is:

- Rapid open-path monitoring of large areas, with 4 km radius, for toxic air pollutants
- Rapid unplanned measurements of fugitive releases anywhere within 4 km radius
- Reduced personnel exposure to harmful gases
- Reduced monitoring costs for large areas
- User friendly operation via simple operator controls
- Easy movement and use in any location, e.g., mountains, cities, etc.
- Does not impact other activities because system is completely self-contained
- Easy setup, e.g., does not use retroreflectors and all equipment is in one location
- Easily understood results given by bar charts showing concentrations and danger levels
- Unattended operation permitted by computer control and automatic recording instruments

Future Activities

The project schedule is shown in Figure 11. Many analyses and calculations on system performance have been completed and this task will be completed by the end of 1996. The system design is virtually completed as is the development of the user graphical interfaces. The program coding for the control computer and the analysis computer has been started and will continue through next April. One AOTF has been fabricated and two more crystals have been grown. The crystals are being fabricated into AOTFs. Several hardware orders have been placed and deliveries will continue through next March. System assembly and testing will start in November and continue through next March. Performance evaluation of the Monitor will then be performed in the Northrop Grumman STC laboratory through June 1997.

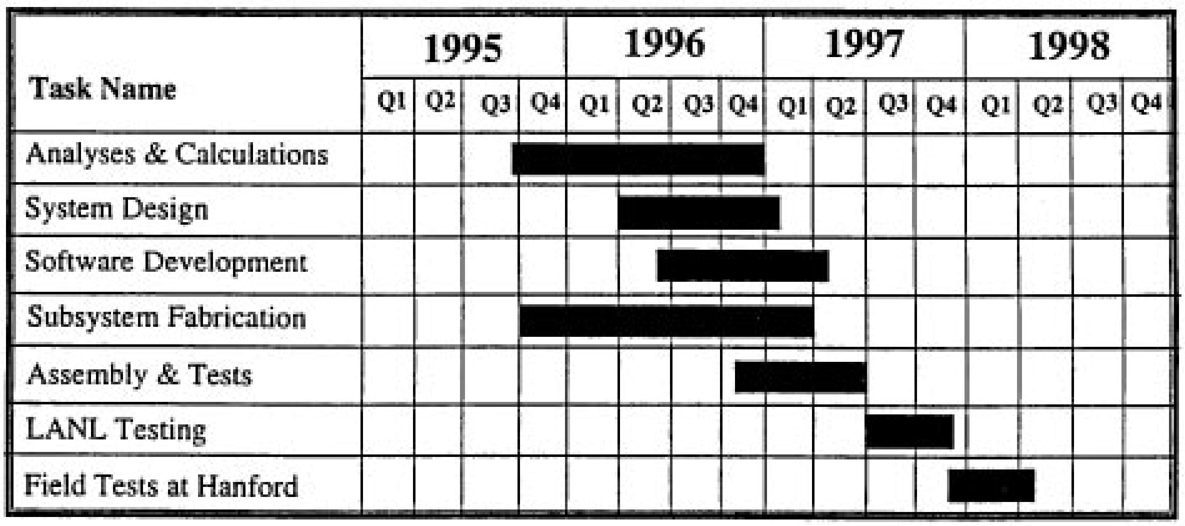

Figure 11. The program schedule includes considerable performance testing.

At the conclusion of the laboratory testing, the TELA Monitor will be installed into a vehicle, probably an RV, and taken to the Los Alamos National Laboratory where there is a secured test range. At this test range, known amounts of trace gases of interest to the program can be emitted into the open air at measured distances from the Monitor. Five months of testing, July through November of 1997, will provide the information needed to fully optimize and evaluate the TELA Monitor.

In December 1997, an environmental impact report will be prepared for testing at the DOE Hanford Site. The Monitor will then be moved from Los Alamos to Hanford. Field tests at the Hanford tank farms will be performed from January through May of 1998. These field tests will evaluate the TELA Monitor under actual operating conditions, including various times of the day and in different kinds of weather. METC personnel will participate in the Monitor testing at both Los Alamos and at Hanford.

Acknowledgments

We gratefully acknowledge the support of the Morgantown Energy Technology Center Contracting Officer's Representative, Jagdish L. Malholtra. We also appreciate J. Huckaby of the Pacific Northwest Laboratory for supplying the list of chemicals detected in the DOE tanks and J. Schultz for helping establish the interaction between Northrop Grumman and the Los Alamos National Laboratory.

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